

The influence of the bulk density on the intergranular properties of $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_Y$ ($0 \leq x \leq 0.01$) ceramics

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Abstract

The influence of the bulk density of $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_Y$ ($0 \leq x \leq 0.01$) ceramics on the intergranular superconducting (SC) properties was studied using the temperature dependence of AC magnetic susceptibility measurements. It was found that the simultaneous variation of the sample's density and the iron impurity concentration does not influence effectively the onset temperature of the superconducting state T_c^{on} . While only increasing of the sample's density shifts the intergranular hysteresis losses peak temperature T_m^J to the lower values which connects with the decreasing of the Josephson magnetic vortices pinning role. It was established that the shielding capability and T_m^J display a plateau with X in the $0.003 \leq X \leq 0.007$ region which is due to the monotonous decrease of the sample's density. It was shown that the shielding capability at the $T = 78K$ for the sample with $3.8g/cm^3$ is two times higher than that for the sample with the density of $5.0g/cm^3$. The possible interpretations of the observed results are discussed.

1 Introduction

About the crucial role of bulk density (ρ) in the formation of SC properties in the oxide alloys was discussed in refs. [1-9]. The bulk density ρ was defined by sintering pressure and by choosing the pressure regime and also by temperature and annealing time of the sintering [1-4].

At the same time, the decrease of the granule sizes and the increase of the ρ and further saturation was observed when the sintering pressure reaches the limiting value [1, 2]. It was shown that the pressing rate is a more critical parameter than the value of sintering pressure in the formation of the SC and structural properties of the obtained samples [5]. In ref. [6] was observed a decrease of the oxidation rate with the increase of density of $YBa_2Cu_{3-X}Fe_XO_Y$ ceramics which makes it difficult to grow a sample with a higher X . An increase of critical current (J_c) and magnetic susceptibility(χ') in the superconducting state and also a decrease of resistivity of the sample in normal state was observed with increasing of ρ in [1,2,7,8]. While in ref. [3] the samples with higher ρ have lower J_c . On the other hand, it was established that the onset temperature T_c^0 of the resistivity appearance increases, and the onset temperature for diamagnetism T_d^{on} decreases with the increasing of ρ in Y- and Bi-based samples [7, 8].

Note, also that the critical transition temperature T_c in Bi-based samples remains unchanged [2], although their density ρ varies in a significantly large range. At the same time the critical temperatures, defined by onset, midpoint and offset of SC transition, depending on sintering pressure (or on density ρ), display nonmonotonic behavior [4, 5]. In $Bi_{3.2}Pb_{0.8}Sr_4Ca_5Cu_7O_Y$ samples the hysteresis losses peak (χ'') temperature T_m^J and onset temperature of diamagnetism (χ') T_d^{on} shifts to the lower values with increasing of their ρ [8].

It was shown [9], that the temperature of complete penetration of high frequency ($\sim 4\text{ MHz}$) electromagnetic field into SC $YBaCuO$ plates T_m^J shifts to the lower value region with increasing of ρ , in contrast with [8]. Thus the analysis of literature data shows that the role of ρ in formation of physical properties of HTSCs is important and it is not clarified unambiguously at present.

In this paper using AC susceptibility measurements we present the results of study of intergranular SC properties of the $YBa_2Cu_{3-X}Fe_XO_Y$ ($0 \leq X \leq 0.01$) ceramics with fixed and variable compositions depending on ρ .

2 Experimental

An AC susceptibility $\chi' - i\chi''$ measurement technique was applied in order to determine the critical parameters of high T_c superconductors. The experimental equipment is described in detail in our previous papers [10, 11]. Note that the sample's temperature changed from 78 to 100 K and was measured within the accuracy of ± 0.1 K.

The samples of about 3 mm in diameter and about 9 mm in height were prepared by the well known ceramic technology. The concentration X of Fe^{57} isotopes which substituted copper atoms were: 0; 0.002; 0.003; 0.005; 0.007 and 0.01 which are subsequently referred as # 0, 1, 2, 3, 4 and 5. For these samples the T_c^{on} was changed between 91 and 92 K, and their bulk density had the following values: 5.29; 5.22; 4.64; 4.71; 3.92 and 5.00 g/cm^3 , respectively. We have also studied another sample #6, containing atom of Fe^{56} isotope with $X = 0.01$ as a substitutional element for copper and having the following parameters: $\rho = 3.8 g/cm^3$; $T_c^{on} = 91.5$ K. The sample's density was defined by their weights and sizes. Note that we do not measure the peak positions of χ'' for sample #5 because they were below 78 K [11].

Note, that in our investigation we also use the shielding capability of the samples. As such a parameter we have chosen the value of χ' at 78 K in arbitrary units.

3 Results and discussions

Fig. 1 shows the temperature dependences of imaginary χ'' (a) and real χ' (b) parts of magnetic susceptibility for the samples with variable composition and different density (curves from 0 to 5). The analogous results are presented in the same fig. (curves 5 and 6) for samples #5 and #6 with the same composition $X = 0.01$ and different densities 5.00 and 3.87 g/cm^3 , respectively.

As is seen in fig.1 with the increasing of X a shift of both the T_m^J peak position on the $\chi''(T)$ curve and the corresponding low temperature inflection point on the $\chi'(T)$ curve to the low temperatures was observed which agrees with our previous results [11]. Such a behavior displays also the samples having $X = 0.01$ with increasing of ρ . At the same time the broadening of the SC transition was observed on both curves $\chi'(T)$ and $\chi''(T)$. The

strong dependence of SC properties versus density is obvious from fig. 1 where the T_m^J for sample #5 was shifted to below the temperature of liquid nitrogen while for sample #6 $T_m^J = 88.8$ K . This behavior agrees with our previous results of T_m^J versus X and it can be explained as follows. The T_m^J decreasing with increasing of X is due to the formation of weak links in the granuls [11,12]. And the observed decrease of T_m^J with increase of sample's density , as assumed in [7,8], may be caused by the reduction of the role of Josephson vortices pinning which is in its turn due to the reduction of the thickness of intergranular layers. It can be mentioned that the high density samples sintered by conventional method, have low oxygen content [6] which enhances the role of inhomogeneities induced in samples by iron doping [11, 12, 13, 14]. Hence, these inhomogeneities lead to the additional broadening of the SC transition in denser samples (fig.1 , curves 3 and 4 ; 5 and 6).

One can also indicate a peculiarity of $\chi'(T)$ curves: the shielding capability (fig. 1. B; 5 ,6 curves) decreases twice with the increase of ρ for the fixed value $X = 0.01$ which contradicts the results in [7,8]. This indicates additionally that the samples with higher density can have an oxygen deficiency [6] which cause the reduction of χ' [15, 16].

Note, that the T_c^{on} versus ρ shows a fluctuation character and ranges in 91 - 92 K interval. Such behavior was found in undoped Y- based samples [4,5] with the difference that the T_c^{on} changes monotonously from 89 to 95 K with increasing of ρ from 3.4 to 5.4 g/cm^3 . The variation of T_c^{on} with ρ according to [4,5] can be explained as follows. The samples with higher density are sintered under the high pressure and have a high concentration of structural defects, created by plastic deformation after the pressure. These defects are probably concentrated near the grain boundaries and reduce strongly the T_m^J , which is observed in our experiments for the #5 sample having higher density in comparison with sample #6. If the accumulated structural defect concentration exceeds some critical value, the sample undergoes from orthorhombic to the tetragonal type of symmetry [4,5]. Hence in order to obtain samples with higher density and better SC properties it required that they undergo special thermal treatment [5].

From the magnetic field dependence of SC transition in sample one can speculate about pinning of magnetic flux lines, which leads to gaining information about the HTSC phenomenon [11,12]. The run of $\chi''(T)$ curves for some applied AC magnetic field amplitudes in sample #3 are presented in

fig. 2. And curves of $T_m^J(h_0)$ for all samples are also presented in fig. 3. The common behavior for all samples is the shift of T_m^J to the lower values with increasing of magnetic field amplitude h_0 . It is typical that all curves $T_m^J(h_0)$ reveal strong and weak region of decreasing of T_m^J which evidences about the existence in samples of "strong" and "weak" pinning centres for Josephson magnetic vortices [11]. From figures 2, 3 one finds that the magnetic field h_0 region for strong decreasing of T_m^J is narrowing with increasing of ρ which agrees with the results obtained from the measurements of high frequency electromagnetic field absorption in Y-based samples [9]. However it should be emphasized that the latter, well agrees with the results of refs. [7,8], but contradicts ref. [9]. The reason for the discrepancy is not obvious, however, it may be attributed to the different techniques for detecting of critical parameters in referred works. For instance, in Y - based samples the onset temperature of diamagnetism T_d^0 decreases [7,8], while the onset temperature of appearance of the electroresistivity, increases with increasing of ρ [7].

As it is known [9,11], the slope of $T_m^J(h_0)$ curve characterizes the pinning force for Josephson vortices and the higher the slope is the lower the pinning force becomes. One can find from fig.3 that for sample #3 with lower iron content and higher ρ in respect to sample #6, the curve $T_m^J(h_0)$ at $h_0 < 0.1$ Oe has a higher slope. Hence, the increase of sample density leads to the decrease of pinning force and/or to decrease of the pinning role of Josephson vortices [7,8].

In #2, 3, 4 samples the iron content X increases monotonously while their densities almost monotonously decrease. Hence the dropping rate of $T_m^J(h_0)$ in these samples consequently decreases. Fig. 4 represents both the dependence of T_m^J for several h_0 and the shielding capability at $T = 78$ K versus iron concentration which clearly describes the role of bulk density in variation of SC properties of samples. It is obvious that these two parameters display the correlative behavior: they sharply drop with initial increasing of X and formate a plateau in $0.003 \leq X \leq 0.007$ region and further decrease when X reaches to 0.01(sample #5). The observed plateau can be explained as follows. In the first two samples the density almost does not change, only the iron concentration X increases which leads to the decrease of T_m^J [11,12]. Further, although the X increases, the ρ almost monotonously decreases which according to [7,8] leads to the decrease of dropping rate of T_m^J versus X and hence causes the formation of a plateau in the above mentioned X region. Further, the simultaneous increase of X and ρ leads to the strong

shift of T_m^J to the temperatures below 78 K [8], as seen from fig. 1 (curve 5). Therefore in fig. 4 the imaginary run of T_m^J for $X = 0.01$ is presented by dotted lines. From this figure one can see that the sample #6, even for relatively higher values of h_0 , has significantly higher values of T_m^J (fig. 4, symbols 4 and 5). This means that in the samples with higher density the T_m^J drops rapidly with increasing X .

4 Conclusion

The analysis of intergranular SC properties of $YBa_2Cu_{3-X}Fe_XO_Y$ ceramics with different X and bulk density allows to draw the following conclusions.

1. The onset transition temperature to the superconducting state T_c^{on} shows a fluctuation character and varies from 91 to 92 K with increasing bulk density and iron concentration.
2. The intergranular losses peak temperature T_m^J decreases rapidly with the increasing of iron content in the samples with a higher density. Partially, in the samples with $X = 0.01$ and higher density the shift of T_m^J to the region below 78 K was observed.
3. The simultaneous increase of iron concentration and bulk density in the samples leads to the rapid decrease of T_m^J . However, a plateau is observed on the $T_m^J(X)$ curves in the $0.003 \leq X \leq 0.007$ region due to the almost monotonous decrease of density in the same region.
4. The shielding capability at 78 K also shows a plateau in the same X region. Besides, it was found that this parameter is about twice lower for the samples with fixed $X = 0.01$ and higher density, which contradicts the results obtained in refs. [7,8]. Such behavior probably may be attributed to the oxygen deficiency in the samples due to the higher bulk density.

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Figure 1: Temperature dependence for real χ' (b) and imaginary χ'' (a) parts of AC magnetic susceptibility for samples. The curves from 0 to 6 correspond to #1, #2, #3, #4, #5 and #6 samples, respectively.

Figure 2: Temperature dependence for imaginary χ'' part of AC susceptibility in sample #3 for applied AC magnetic field amplitudes of h_0 (mOe); 1 - 10; 2 - 30; 3 - 100; 4 - 250.

Figure 3: Dependence of intergranular losses peak temperature T_m^J versus AC magnetic field amplitude h_0 in samples. The curves from 0 to 6 correspond to #1, #2, #3, #4 and #6 samples, respectively.

Figure 4: Dependence of intergranular losses peak temperature T_m^J for several values of h_0 (1, 2, 3, 4, 5 curves) and shielding capability at 78 K (6) versus iron concentration X. 1, 2 and 3 - $T_m^J(X)$ curves for applied AC magnetic field amplitudes h_0 (Oe): 0.025, 0.1 and 0.3, respectively; 4 and 5 - T_m^J values in sample #6, respectively for $h_0 = 0.1Oe$ and $h_0 = 0.3Oe$.